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Using Nuclear Resonance Fluorescence to Probe Nuclear Materials

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Abstract

We explore the feasibility of using Nuclear Resonance Fluorescence (NRF) to determine Pu mass in spent fuel with a photon source. Knowing the amount of Pu in spent fuel is important for domestic and international safeguards. We use an analytical model and Monte-Carlo simulations to determine counting rates as a function of ^{239}Pu concentration in a UO_2 fuel matrix. For this paper, we focus on quasi-monoenergetic photon sources to excite NRF states and absorption detectors to look at the absorption spectrum after passing through the UO_2 fuel matrix.

Introduction

In the past two decades and moreso in the past few years, there is an increasing uncertainty in the management of nuclear materials, especially from the international point-of-view. Subsequently, proliferation of nuclear materials is a topmost concern. Programs by national security and international safeguards communities have focused on a myriad of projects to detect nuclear materials in an effort to defeat proliferation of nuclear material. Some of these programs focus on passive detection while others focus on active interrogation with neutron or photon sources ([Pru2006], [Weh2006], [Ber2005], [Sla2005], [Shu2004], [Bor2003], [Jon2002], [Gma1999], etc…). An emerging game-changer for gamma-ray interrogation methods is Nuclear Resonance Fluorescence (NRF). Besides detecting nuclear materials in bulk quantities, there is an urgent need to detect dilute nuclear materials within a larger matrix. For example, from a safeguards point-of-view, it is important to detect and quantify the amount of ^{239}Pu in nuclear fuel assemblies (UO_2). Knowing the concentration of Pu in a fuel assembly is important for two reasons; first, Pu concentrations (or any other isotope generated from nuclear processes within a reactor) can yield important information about reactor performance. Second, Pu assay is important to detect Pu diversion at critical points after the removal of nuclear fuel assemblies. NRF is a physical process that is uniquely able to perform such assay.

NRF is a photonuclear reaction that occurs when a photon excites a nucleus to a certain energy level. These levels are resonances with very narrow widths. The levels are one of two different types of dipole excitations, an electric dipole excitation (Octupole-Quadrupole vibration) or a magnetic dipole excitation (scissors mode). NRF states in actinides have been shown to have relatively large cross-sections (*c.f.* [Ber2008] *and references therein*). Since each nucleus has a unique NRF signature, detection systems and assay technologies, based on NRF, will be isotopically sensitive. NRF is an inelastic process that does not transmutate the material being probed. That is to say that NRF is a non-destructive method to detect and assay nuclear materials. Therefore, NRF-based assay systems can detect ^{239}Pu in nuclear fuel assemblies (UO_2) without destroying the fuel assembly or altering the isotopic content. Much work is being done to show the effectiveness of NRF in detection roles, assay roles, etc… by many researchers who are looking at NRF-based assay systems including those of us at LLNL. This paper presents the historical and ongoing efforts at LLNL to exploit NRF for detection and assay; however, for the purposes of this paper, we will focus on Pu assay in nuclear fuel matrices.

Detection Protocols

There are two basic ways to detect NRF signals from a gamma-ray illuminated sample. First is a direct measurement of the emitted photons from the photo-excitation from resonant photons hereby dubbed a reflection detector. The other method is an absorption type detector that is sensitive to the absorbed photon flux of the resonant photons (*c.f.* [Met1959]) dubbed a transmission detector. It has been shown that for NRF-based detection of special nuclear materials in heavy shielding the transmission method was generally better (*c.f.* [Joh2010]). Some of us (See [Hag2009]) studied the effects of using various detection methods for NRF-based systems using quasi-monoenergetic photon sources for shielded materials. (It is important to point out that the results in [Hag2009] are sensitive to the choice of experimental input parameters.) Our results indicated that the transmission method, described above, has the highest rate of accuracy in detecting low quantities of nuclear materials of interest in heavily shielded configurations. In a safeguards application where assay is the focus, the Pu concentrations sought after can be quite low (a few percent) and is in a heavily shielded matrix of a fuel assembly. If we use our detection results in [Joh2010] as guidance for the assay problem then it stands to reason that the transmission method, in conjunction with quasi-monoenergetic photon sources, is *a priori*, the best method for NRF-based safeguards assay. Another reason for using the transmission method is reduced backgrounds. For reflection mode, the detectors are facing the fuel assembly. These assemblies, especially spent fuel, contribute large backgrounds because of the high radioactivity of the fuel material. This was not the case for the study by Johnson *et al.* [Joh2010], where the focus was on detecting special nuclear material in cargo containers where the only contribution to the background was low-level normal background. The difference between assay and detection will come up many more times throughout this paper and it is important that this distinction is clear. Additional backgrounds come from beam-correlated events. The benefit of using a quasi-monoenergetic photon source, is that the dominant beam-correlated backgrounds are the elastic channels. The elastic backgrounds have a low count rate and have little effect on the signal-to-noise ratio (*c.f.* [Hag2009], [Sch1980]).

There are different ways to construct a transmission detector. The study by Johnson *et al*., [Joh2010] focused on incorporating high-resolution gamma-ray detectors to make spectroscopic determinations on the presence of nuclear material. Another method, under study at LLNL, uses a low-resolution detection system, but has the advantage of assaying at higher rates (orders of magnitude). For the purposes of this paper, we will focus on the high-resolution transmission detection method, since we can immediately verify our results with prior experimental data.

Assay is much different than detection alone. For detection, the questions being answered are: is there a threat material present? What is the uncertainty of detecting the presence of the threat material and conversely what is the uncertainty that the threat is not present? The latter question directly speaks to the probability of false positives and false negatives. For assay, the questions answered by NRF are: how much of the [^{239}Pu] material is present? And to what level of certainty is associated with that determination? This is addressed further in the next section.

NRF-Based Assay Estimates

There is much to do to show that NRF can play a role in fuel assay. The first exercise is to show estimates on the performance of NRF-based assay systems. Often, it is useful to exploit analytical models to make estimates. More often than not, however, analytical estimates are incorrect because there are large higher-order corrections that such simple models cannot handle. It was shown that for the NRF-based detection setup we described in [Joh2010], there is a simple analytical model that works quite well. The effectiveness of the model comes about from two aspects. First, we are examining quasi-monoenergetic photon sources whose dominant beam-correlated backgrounds are

elastic and can be easily calculated [Sch1980 *and references therein*]. Second, we are using high-resolution spectroscopy and can be easily separated from other contamination signals. Given the success of our analytical model in [Joh2010] we make the following count rate estimates for various concentrations of ^{239}Pu in a UO_2 fuel matrix. These estimates are shown as the black curve in figure 1.

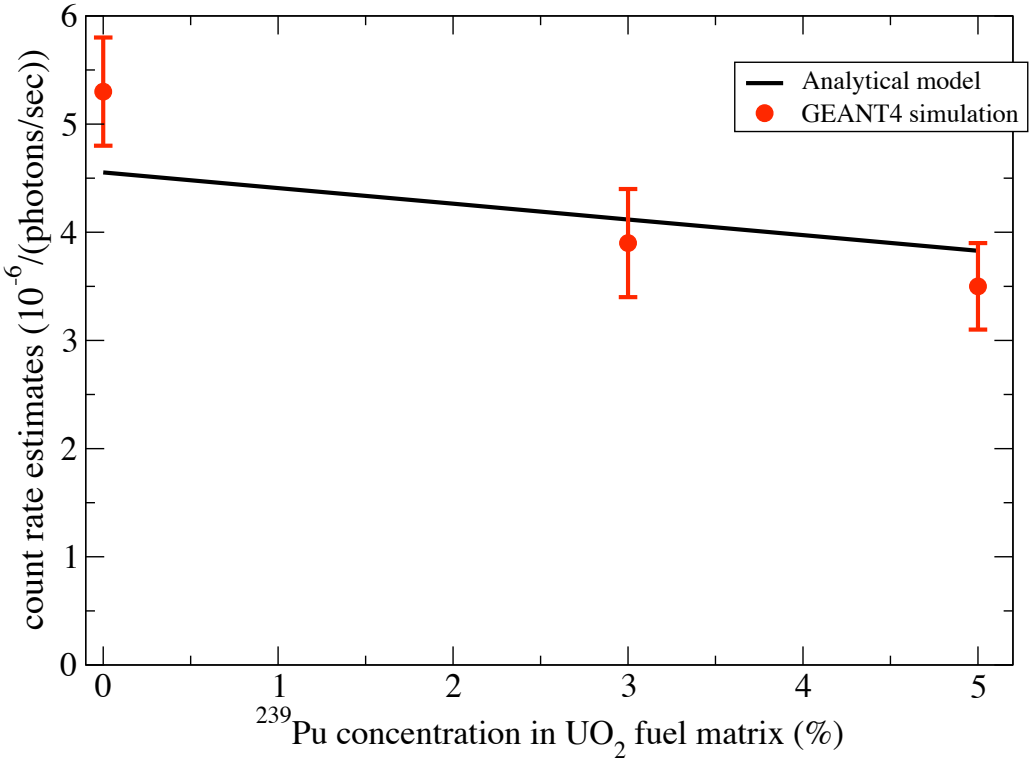

Figure 1: Count rate estimates (vs ^{239}Pu concentration) for NRF-based assay using a transmission detector and a narrow-band gamma-ray source. The black line was generated with a simple analytical model used for a prior effort [Joh2010] and adopted for here. The data are MC simulations made with GEANT4 (see text for details).

For the results in Fig. 1 we assumed a uniform distribution of ^{239}Pu in the UO_2 fuel matrix. We also assume a uniform distribution of the UO_2 fuel material (i.e. no gaps in the fuel assembly). We use a 10 cm thick fuel assembly, which is comparable to actual fuel assemblies. We also assumed a monoenergetic source with a bandwidth comparable to the Doppler-broadened natural linewidth of the resonances. It is important to note that the above curve is a function resonant energy, resonant

strength, source bandwidth, source brightness, detector type, collimation, background shielding, material properties including isotopic distributions within the fuel assembly, and the overall geometry of the fuel assembly. Therefore, the above will change as a result of varying any one of these parameters. The source brightness will determine the time it takes determine the amount of Pu in a spent fuel assembly. These effects of the above model to changes in the parameters must be studied with simulations and validated with measurements and is part of our ongoing studies.

Figure 1 can be viewed as a curve that can be used (for the parameters described above) to determine the concentration of ^{239}Pu as a function of integrated counts. It should be clear to the reader, that if counting statistics are reduced the uncertainties will increase and the accuracy for determining the ^{239}Pu concentration will decrease. From the analysis of our model, we have deduced that the precision to which the concentration can be determined (uncertainty in the ^{239}Pu concentration) is almost 7 times more than the uncertainty of the NRF signal. It is possible that the linear model in figure 1 does not contain all the necessary microscopic details (such as second-order scattering processes), especially when more realistic input parameters are put into the model. From our preliminary studies, we know that the curve in figure 1 changes as a function of bandwidth (and most likely other parameters as well). This is part of our ongoing investigation and will be the focus of benchmarking measurements.

To improve the robustness of the NRF-based assay estimates, it is imperative to perform realistic Monte-Carlo (MC) simulations. In the above figure (Fig. 1), we have superimposed three MC estimates generated with GEANT4 [GEANT4]. The large uncertainties in the data points in figure 1 are due to limited statistics. The simulations were only run to about 10% statistics, due to the limitations posed by performing such calculations on a laptop. The results of the simulations are in good agreement with the analytical model with a chi-squared per degree-of-freedom of 0.989. (The NRF process is not included in the usual GEANT4 package. We, at LLNL, are developing an NRF capability for GEANT4. Prior work to implement NRF in GEANT4 has met with marginal success [Jor2007]. The LLNL effort is an independent project to circumvent the unknown problems experienced in [Jor2007] and to be compatible with developing NRF databases.)

The agreement of the MC data with our experimentally proven analytical model [Joh2010] indicates the success, so far, of the NRF implementation in GEANT4. Additionally, we have matched the NRF count rate for the measured 2144-keV resonance in ^{239}Pu [Ber2008] with a chi-squared per degree-of-freedom of 1.11. This is much better than the absolute results given in [Jor2007] for other isotopes. Validation of NRF for other resonances and isotopes are ongoing. The implementation of NRF in GEANT4 will be used with the other MC simulation programs (e.g. MCNP [MCNP] and COG) to test the feasibility of using NRF to assay fuel materials as well as other NRF applications. A similar effort is underway at Los Alamos National Laboratory to implement NRF in MCNP. Work performed at Brookhaven National Laboratory is ongoing to create a database that can be used by the various MC programs for NRF-based applications.

We have performed MC simulations with an LLNL-based MC code called COG. NRF was developed for COG as part of a study to look at NRF-based imaging and assay of nuclear fuel rods. The results of that study are published in [Joh2010nd]. It was determined as a result of the COG calculations that isotopic defects on a sub-mm scale that range from 0.2% to 25.0% could be characterized by NRF. It is a small step in reasoning to realize that if NRF can be used to map small isotopic defects, it can be used to look for larger scale deviations within fuel assemblies. This indicates that NRF can be used to isotopically map fuel assemblies. Although assay and imaging are mutually exclusive, they can be combined to expand the usefulness of NRF-based applications for safeguards.

Conclusion

In summary, we have estimated that NRF can measure a homogenous distribution of Pu material in a UO_2 fuel assembly with both MC simulations and analytical calculations. It has been indicated that the estimates for NRF-based detection, assay etc… are sensitive to input parameters (e.g. source type, detector types, collimation, resonance energy/width, etc…). This implies that the above studies need to be extended for a variety of input parameters. It has been discussed that NRF-based detection is vastly different from NRF-based assay and is also different from imaging. We have shown, with MC simulations, NRF can be used to assay and map isotopic defects in a standard fuel rod. We have focused on quasi-monoenergetic sources and transmission detection, but it is important to note that for safeguards, there may be applications where either or both of those approaches would be best. It is critically important that computational estimates be followed with benchmarking measurements to validate models.

Acknowledgments

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